I

JC13 Rec'd PCT/PTO 27 APR 2005

Claims

Substituted 6-(2-halogenphenyl)-triazolopyrimidines of formula I

42

10

in which

Hal is halogen;

15

 L^1, L^3 independently denote hydrogen, halogen, or $C_1-C_4-alkyl;$

L² is hydrogen, halogen, C_1-C_4 -haloalkyl, or NH₂, NHR^b, or N(R^b)₂,

20

25

Rb is C_1-C_8 -alkyl, C_3-C_{10} -alkenyl, C_3-C_{10} -alkynyl, C_1-C_6 -haloalkyl, C_3-C_6 -haloalkenyl, C_3-C_6 -haloalkynyl, C_1-C_8 -alkoxy- C_1-C_8 -alkyl, C_1-C_8 -alkylthio- C_1-C_8 -alkyl, C_3-C_{10} -cycloalkyl, or C(=0)-A, in which

A is hydrogen, hydroxy, C_1-C_8 -alkyl, C_1-C_8 -alkoxy, C_1-C_6 -halogenalkoxy, C_1-C_8 -alkylamino or $di-(C_1-C_8-alkyl)$ amino;

wherein at least one from L^1 , L^2 , and L^3 is not hydrogen;

X is halogen, cyano, C_1 - C_6 -alkyl, C_1 - C_6 -alkoxy, C_1 - C_6 -haloalkoxy or C_3 - C_8 -alkenyloxy.

35 R^1 denote C_1-C_{10} -alkyl, C_2-C_{10} -alkenyl, C_2-C_{10} -alkynyl, or C_4-C_{10} -alkadienyl, C_2-C_{10} -haloalkenyl

wherein \mathbb{R}^1 may be unsubstituted or may carry one to three groups \mathbb{R}^a ,

40

Ra is cyano, nitro, hydroxyl, $C_1-C_6-alkyl$, $C_3-C_6-cyclo-alkyl$, $C_1-C_6-alkoxy$, $C_1-C_6-alkyl$ thio, $C_1-C_6-alkyl$ amino, $di-C_1-C_6-alkyl$ amino, C_2-C_6-alk enyl, C_2-C_6-alk enyloxy, C_2-C_6-alk enyl, C_3-C_6-alk enyloxy, C_3-C_6-alk enyloxy, C_3-C_6-alk enyloxy,

or C_1-C_4 -alkylenedioxy;

R² is hydrogen;

- 2. Compounds of formula I according to claim 1, in which
- 5 R^1 is straight chained or branched C_2 - C_6 -alkenyl, C_1 - C_6 -alkyl.
 - 3. Compounds of formula I according to claim 1 or 2 in which X is halogen.

10

- 4. Compounds of formula I according to any one of claims 1 to 3 in which the 6-(2-halogenphenyl)group represents one of the following moieties:
- 2,3,5-trifluorophenyl, 2-F,4-CF₃-phenyl, 2-F,5-CH₃-phenyl,
 2-Cl,4-F-phenyl, 2-F,4-Cl-phenyl, 2-F,4-Br-phenyl, 2-Cl,4-Brphenyl, 2,3-difluorophenyl, 2,4-difluorophenyl, 2,4,5-trifluorophenyl, 2,3,4-trifluorophenyl, 2-F,4-NHC(O)CH₃-phenyl,
 2-Br,3,5-difluorophenyl, 2-F,4-NO₂-phenyl, and
 20 2-Cl,4-NO₂-phenyl.
 - 5. A process for the preparation of compounds of formula I as defined in claims 3 and 4 which comprises reacting 5-amino-1,2,4-triazole

25

with 2-phenyl-substituted malonic acid ester of formula II,

30

35

wherein Hal, L^1 , L^2 , and L^3 are as defined in formula I, and R denotes $C_1\text{-}C_6\text{--alkyl}$, under alkaline conditions, to yield compounds of formula III,

40

which are subsequently treated with a halogenating agent to give 5,7-dihalogen-6-phenyl-triazolopyrimidines of formula IV

15

44

in which Y is halogen, and which is reacted with an amine of formula \boldsymbol{V}

in which \mathbb{R}^1 and \mathbb{R}^2 are as defined in claim 1 to produce compounds of formula I, as defined in claim 1.

6. A process for the preparation of compounds of formula I according to claim 1 wherein X is cyano, C_1-C_{10} -alkoxy, or C_1-C_6 -haloalkoxy, which comprises reacting 5-halogen-triazolopyrimidine of formula I',

20

R¹
N R²
L³
L
1

I

25

wherein Y is halogen, with compounds of formula VI,

which are, dependent from the value of X' to be introduced, an anorganic cyano salt, an alkoxylate, haloalkoxylate or an alkenyloxylate, resp., wherein M is ammonium-, tetraalkylammonium-, alkalimetal- or earth metal cation, to produce compounds of formula I.

7. Intermediates of formulae II, III, and IV as defined in claim 5, in which the 6-(2-halogenphenyl) group represents one of the following moieties:

2,3,5-trifluorophenyl, 2-F,4-CF₃-phenyl, 2-F,5-CH₃-phenyl, 2-Cl,4-F-phenyl, 2-F,4-Cl-phenyl, 2-F,4-Br-phenyl, 2-Cl,4-Br-phenyl, 2,3-difluorophenyl, 2,4,5-trifluorophenyl, 2,3,4-trifluorophenyl, 2-F,4-NHC(O)CH₃-phenyl, 2-Br,3,5-difluorophenyl, 2-F,4-NO₂-phenyl, and 2-Cl,4-NO₂-phenyl.

45

35

- 8. A composition suitable for controlling phytopathogenic fungi, comprising a solid or liquid carrier and a compound of the formula I as claimed in claim 1.
- 5 9. A method for controlling phytopathogenic fungi, which comprises treating the fungi or the materials, plants, the soil or the seed to be protected against fungal attack with an effective amount of a compound of the formula I as claimed in claim 1.

10. Substituted 6-(2-halogenphenyl)-triazolopyrimidines of formula I

in which

20

30

35

45

Hal is halogen;

 L^1, L^3 independently denote hydrogen, halogen, or C_1 - C_4 -alkyl;

25 L^2 is hydrogen, halogen, C_1-C_4 -haloalkyl, or NH₂, NHR^b, or N(R^b)₂,

Rb is C_1 - C_8 -alkyl, C_3 - C_{10} -alkenyl, C_3 - C_{10} -alkynyl, C_1 - C_6 -haloalkyl, C_3 - C_6 -haloalkenyl, C_1 - C_8 -alkoxy- C_1 - C_8 -alkyl, C_1 - C_8 -alkylthio- C_1 - C_8 -alkyl, C_3 - C_{10} -cycloalkyl, or C(=0)-A, in which

A is hydrogen, hydroxy, C_1-C_8 -alkyl, C_1-C_8 -alkoxy, C_1-C_6 -halogenalkoxy, C_1-C_8 -alkylamino or $di-(C_1-C_8$ -alkyl) amino;

wherein at least one from L^1 , L^2 , and L^3 is not hydrogen;

is halogen, cyano, C_1-C_6 -alkyl, C_1-C_6 -alkoxy, C_1-C_6 -haloalkoxy or C_3-C_8 -alkenyloxy.

R¹ and R² together with the interjacent nitrogen atom represent a saturated or partially unsaturated 5- or 6-membered heterocycle, containing one to four nitrogen atoms or one to three nitrogen atoms and one sulfur or

15

oxygen atom, which ring may be substituted by one to three R^a radicals;

Ra is cyano, nitro, hydroxyl, C₁-C₆-alkyl, C₃-C₆-cyclo-alkyl, C₁-C₆-alkoxy, C₁-C₆-alkylthio, C₁-C₆-alkylamino, di-C₁-C₆-alkylamino, C₂-C₆-alkenyl, C₂-C₆-alkenyloxy, C₂-C₆-alkynyl, C₃-C₆-alkynyloxy, or C₁-C₄-alkylenedioxy;

10 11. Compounds of formula I according to claim 10, in which

 ${\rm R}^1$ and ${\rm R}^2$ together with the interjacent nitrogen atom represent a heterocyclic ring with 5 or 6 carbon atoms being optionally substituted with one or two ${\rm C}_1{\rm -C}_4{\rm -alkyl}$ groups.

- 12. Compounds of formula I according to claim 10 or 11 in which R¹ and R² together with the interjacent nitrogen atom represent a 5- or 6-membered heterocyclic ring being optionally substituted with one or two methyl groups.
 - 13. Compounds of formula I according to any one of claims 10 to 12 in which X is halogen.
- 25 14. Compounds of formula I according to any one of claims 10 to 13 in which the 6-(2-halogenphenyl)group represents one of the following moieties:

2,3,5-trifluorophenyl, 2-F,4-CF₃-phenyl, 2-F,5-CH₃-phenyl,
2-Cl,4-F-phenyl, 2-F,4-Cl-phenyl, 2-F,4-Br-phenyl, 2-Cl,4-Brphenyl, 2,3-difluorophenyl, 2,4-difluorophenyl, 2,4,5-trifluorophenyl, 2,3,4-trifluorophenyl, 2-F,4-NHC(O)CH₃-phenyl,
2-Br,3,5-difluorophenyl, 2-F,4-NO₂-phenyl, and
2-Cl,4-NO₂-phenyl.

15. A process for the preparation of compounds of formula I as defined in claims 13 and 14 which comprises reacting 5-amino-1,2,4-triazole

40 NH NH

45

35

10

15

30

with 2-phenyl-substituted malonic acid ester of formula II,

wherein Hal, L^1 , L^2 , and L^3 are as defined in formula I, and R denotes C_1 - C_6 -alkyl, under alkaline conditions, to yield compounds of formula III,

which are subsequently treated with a halogenating agent to give 5,7-dihalogen-6-phenyl-triazolopyrimidines of formula IV

in which Y is halogen, and which is reacted with an amine of formula V

$$R^1$$
 N-H V

in which R^1 and R^2 are as defined in claim 10 to produce compounds of formula I, as defined in claim 10.

16. A process for the preparation of compounds of formula I according to claim 10 wherein X is cyano, C₁-C₁₀-alkoxy, or C₁-C₆-haloalkoxy, which comprises reacting 5-halogen-triazolo-pyrimidine of formula I',

wherein Y is halogen, with compounds of formula VI,

M-X' VI

which are, dependent from the value of X' to be introduced, an anorganic cyano salt, an alkoxylate, haloalkoxylate or an alkenyloxylate, resp., wherein M is ammonium-, tetraalkylammonium-, alkalimetal- or earth metal cation, to produce compounds of formula I.

17. A composition suitable for controlling phytopathogenic fungi, comprising a solid or liquid carrier and a compound of the formula I as claimed in claim 10.

18. A method for controlling phytopathogenic fungi, which comprises treating the fungi or the materials, plants, the soil or the seed to be protected against fungal attack with an effective amount of a compound of the formula I as claimed in claim 10.